SIMULATION STUDIES OF DIFFUSION-RELEASE AND EFFUSIVE-FLOW OF SHORT-LIVED RADIOACTIVE ISOTOPES

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Abstract

Simulation studies with computer models offer cost effective methods for designing targets and vapor transport systems at Isotope Separator On-Line (ISOL)-based radioactive ion beam facilities. A finite difference code, *Diffuse II*, was developed at the Oak Ridge National Laboratory (ORNL) for studying diffusion-release of short-lived ion species from the three principal target geometries; results derived by use of the code are in close agreement with analytical solutions to Fick's second equation. A Monte-Carlo code, *Effusion*, was developed to address issues related to the design of fast vapor transport systems. Results, derived by the use of *Effusion* closely agree with experimental measurements. These codes and their applications are discussed in this article

INTRODUCTION

Several ISOL-based facilities, including the Holifield Radioactive Ion Beam Facility (HRIBF) [1], have been constructed for production and acceleration of short-lived radioactive ion beams (RIB)s for use in nuclear physics and nuclear astrophysics experiments. Intensities of short-lived species, generated by this technique, are limited by decay losses associated with times for diffusion release from target materials and for effusive flow from the target to the ion source. For example, time required for diffusion of particles from a 5μm thick, planar-geometry, solid target, may range from a few seconds to several hundred hours while the effusive flow times for particles through transport systems, depend on the mass of the species, the porosity of the target material, temperature, geometry and size of the vapor transport system and the chemistry between the species and materials of construction of the system. In order to provide useful beam intensities of short-lived isotopes, it is imperative to minimize delay times for these limiting processes by optimally designing both production targets and vapor transport systems.

Following diffusion–release from the production target, particles must be transported through a target matrix into a vapor transport system that is connected to an ion source where a fraction of the arriving particles are ionized and extracted for acceleration to research energies. Time delays for diffusion usually are longer than those for effusive-flow for noble gas and electropositive elements. However, for electronegative elements with appreciable enthalpies of adsorption, the delay times may be reversed. Vapor transport systems can be optimized in terms of geometry and size to minimize the average distance traveled between the production target an ion source and by choice of refractory materials of construction with low enthalpies of adsorption.

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DIFFUSION

Thermal diffusion is the process in which particles are transported from one region to another through random molecular motion. The diffusion process for a short-lived isotope of concentration C and decay constant λ can be described by Fick's second equation [2]:

$$\frac{dC}{dt} = D \cdot \nabla^2 C + S - \lambda \cdot C \cdot e^{-\lambda \cdot t} \qquad (1)$$

where D is the diffusion coefficient and, S is the production rate of the specie. Diffusion coefficient D depends on the physical and chemical properties of the binary diffusion couple, i.e., target material and specie as well as target temperature. As noted from Eq. 1, the rate of diffusion-release of an isotope from a given target material depends on the production rate, the lifetime of the isotope, diffusion coefficient and dimensions of target material. In order to minimize decay losses, it is important to choose highly refractory production target materials that have large diffusion coefficients and to fabricate targets with dimensions commensurate with fast release of the product species.

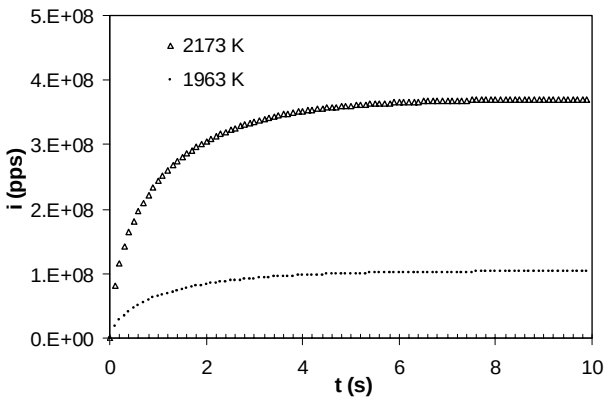

Figure 1. Release of Cl^{34} ($\tau_{1/2}$ =1.53 s) from films of CeS. Film thickness: 3 μm; At 1963 K: D = 4.4×10^{-10} cm^2/s; at 2173 K: D = 5.0×10^{-9} cm^2/s); Generation rate: 1.1×10^9 pps.

Eq. 1 can be solved analytically for the three principal geometries (planes, cylinders and spheres) to extricate time-release information for short-lived species, provided that D is known. However, numerical methods, offer a general approach for obtaining diffusion release-time from arbitrary geometry. The finite difference code, *Diffuse II*, was developed for solving binary diffusion couple problems for arbitrary target-geometries [3]. *Diffuse II* is benchmarked against analytical solutions to Eq. 1 with differences < 1%. Since the finite difference method can be applied to arbitrary boundary and initial

condition problems, the code can be used to design targets with optimum production/release rate properties.

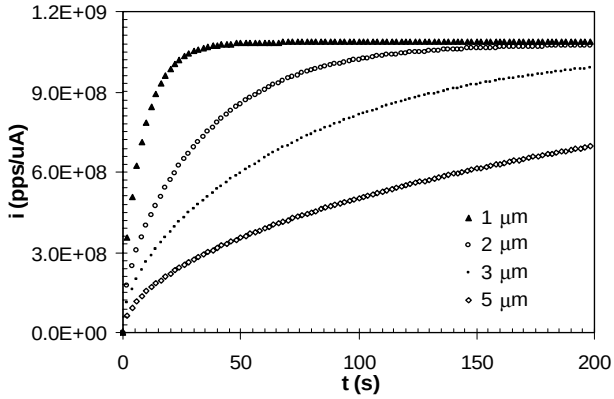

Figure 2. Release of Cl^{34} ($\tau_{1/2}$ = 32 m) from various thickness films of CeS, at 1963 K: D = 4.4 $\times 10^{-10}$ cm^2/s; Generation rate: 1.1×10^9 pps/μA.

Figures 1 and 2, respectively, compare release rates of Cl^{34} ($\tau_{1/2}$ =1.53 s) and Cl^{34m} ($\tau_{1/2}$ =32 m) from various thickness CeS targets irradiated with 50 MeV, 1 μA proton beams. As noted, it is desirable to design targets with high limiting temperatures and with as short as practically achievable diffusion lengths [4].

EFFUSIVE FLOW

To reduce effusive-flow times, we can minimize the distance traveled per particle from the target to the ion source as well as the residence times of particles during adsorption. Careful consideration is given to the geometry and size of the system in addition to choice of refractory material of construction with low enthalpy of adsorption for the isotope of interest.

From vacuum conductance theory, the characteristic evacuate time for particles passing through a straight tube with one end closed under molecular flow conditions with no enthalpy of adsorption, is [5]:

$$t = \frac{4V}{\pi^2 C} \qquad (2)$$

where, V is the volume, and C is the conductance of the tube. For simple geometries, vacuum conductance theory is accurate for computation of flow rates but the accuracy for complex geometries, such as required for RIB vapor transport systems, is quite low. Hence, a more accurate method is required to estimate effusive-flow delay times through arbitrary geometry target/ion source systems. We developed an Monte-Carlo code, *Effusion*, and it closely replicates experimentally measured results [6]. *Effusion* makes use of the GEANT4 toolkit [7] to simulate effusive-flow of particles through a complex 3D target/ion source system.

Effusive-flow of particles from target surfaces through a simple transport system into the ionization chamber of an ion source can be described with vacuum conductance theory, provided that particle sticking times on surfaces are negligible. Figure 3 displays a cross section of a vapor transport system that couples ISOL targets to an electron impact ion source at the HRIBF. The theory is applied to the system by dividing it into various tubular connected volumes. Eq.2 can then be applied to estimate the evacuation time t with which may directly estimate the average distance l traveled per particle in the system from:

$$l = \frac{4}{3} v \cdot t \qquad (3)$$

where, v is the average Maxwell velocity: $(8k_B T/\pi M)^{1/2}$, of a particle of mass M, and T is the absolute temperature [8,9].

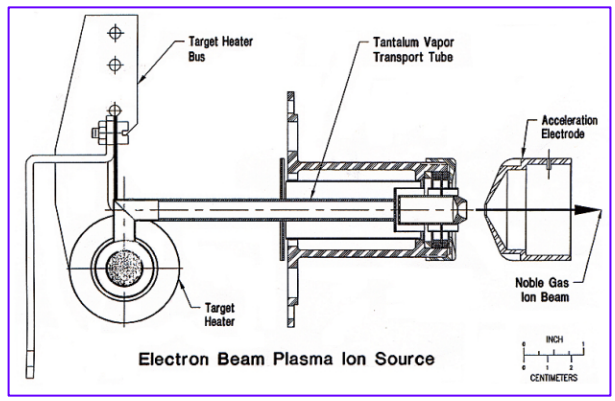

Figure 3. Schematic drawing of a target/ion source system used at the Holifield Radioactive Ion Beam Facility.

However, estimations of effusive flow rates based on the conductance theory are limited to simple connected system, it may not be applied to the generic case which may involve very complex geometries. Also, the theory cannot estimate the affects of surface adsorption through a given system. More accurate and detailed effusive-flow information can be determined either from experimental measurements or by the use of an experimentally vetted Monte-Carlo simulation, such as *Effusion*.

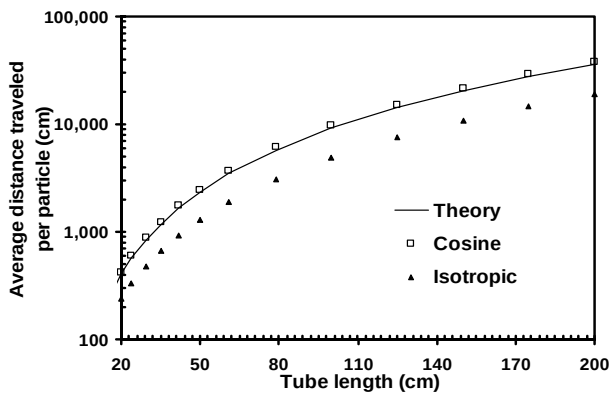

Figure 4. Distance traveled per particle in long tubes. Simulation of cosine reflections and theory is within 4%

Specular reflection, isotropic distribution and cosine distribution models have been investigated with *Effusion*. The code has been validated against vacuum conductance theory for simple geometry and more importantly, against accurate experimental measurements of very complex target/ion source systems.

Simulations of effusive-flow through serial-, parallel- and high conductance- connected vapor transport systems [8,9] were compared with measured values. They agree within 2% of whenever isotropic re-emission distributions about surface normals are assumed, while agreement between experiment and conductance theory is only 20% due to the complexity of each system. But simulation results, using the cosine re-emission distribution model, closely agree with conductance theory for very simple geometries where conductance theory been proved to be consistent with measurement. Figure 4 shows simulation results compared against conductance theory for very long tubes. As noted, differences between simulations with the cosine re-emission distribution model and conductance theory are less than 4%.

Simulations obtained from *Effusion*, using the cosine re-emission distribution model agree with conductance theory and are therefore, accurate for very long tubes. However, simulation results for short tubes do not agree with the theory due to tube end effects that cannot be accurately represented by analytical approximations but are accurately accounted for in Monte-Carlo simulations. Also in the real case where the transport systems are constructed from short tubes, surfaces irregularities have strong influences on effusive-flow through such systems. In the real scenario, surfaces are not atomically smooth and the surface normals are canted in many different angles so that particle ejection appears to be isotropic even for particle re-emission in a cosine distribution. This scenario possibly explains why *Effusion* simulation results obtained with the isotropic re-emission distribution agree with experimental measurements for target/ion source systems while those obtained with the cosine re-emission distribution agree with the conductance theory.

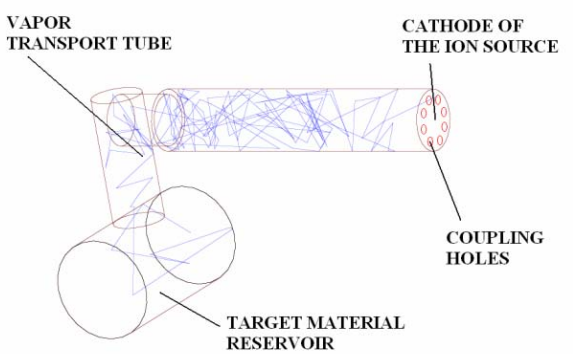

Figure 5. Monte-Carlo simulation of particle transport in the HRIBF target/ion source system

Simulations with *Effusion* code show that the number of particle collisions with the walls is proportional to the average distance traveled per particle through the system. And graphic display of particles trajectories through a system offer a means for detecting points where problems occur and thus, facilitate the design of vapor transport systems with minimum transport time properties. Figure 5 displays particle trajectories within the HRIBF vapor transport system, simulated with the code. The simulation studies show that the longest delay times are caused by the coupling holes in the cathode that allow particle transport into the anode structure of the source. The next element that restricts flow is the vapor transport tube that connects the target to the ion source.

A close-coupled vapor transport has resulted from these studies in which the target is moved forward so that particles from the target travel directly into the center of the anode structure [10]. This arrangement eliminates coupling holes and the vapor transport tube that connects the target material reservoir to the ion source. According to computer simulations, the close-coupled system will reduce particle effusive- flow time by two orders of magnitude, thus, enabling the rapid transport of short-lived isotopes to the ion source. However, the concept has yet not been experimentally evaluated.

CONCLUSIONS

Computer codes have been developed for studies of the diffusion-release and effusive-flow processes, which, in combination, limit ion beam intensity at ISOL facilities. The code *Diffuse II* enables the study of the release properties of short-lived species from arbitrary geometry target and thus, enables the design of targets with optimum release properties. A very powerful Monte-Carlo code, *Effusion*, has been developed that agrees closely with conductance theory for simple systems and with experimental measurements for very complex systems. *Effusion* has been utilized to estimate the effusive-flow properties of a new, concept vapor transport system that reduces delay times by ~ two orders of magnitude.

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